DEVELOPING EXPERIMENTAL CAPABILITY FOR INVESTIGATION OF FREE RADICAL PROCESSES UNDER LIGHT WATER REACTOR OPERATING CONDITIONS

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Water radiolysis at extreme conditions of high temperature and high pressure (HTHP) is an important issue in a number of areas in nuclear reactor technology, especially in the high temperature corrosion of structural materials in the primary circuit of Light Water Reactors (LWRs), which is noticeably enhanced by the oxidizing radical products of water radiolysis. Mitigation of the detrimental effects of water radiolysis on reactor's structural components promotes the long-term viability of water-cooled nuclear systems. In addition, the choice of promising materials for the next generation of nuclear reactors will rely on the fundamental understanding of the radiation chemical processes in water under extreme conditions of high heat, pressure and mixed radiation fields.

Direct examination of the free radical processes in nuclear reactor cores is extremely difficult due to the intense mixed radiation fields and HTHP [1]. One way to circumvent these limitations is to design an experiment in which water (or aqueous solution) under study is irradiated in the high temperature/high pressure conditions using conventional radiation sources. In my talk I will describe the design of experimental capability dedicated to the study of radical processes arising from the radiolysis in HTHP flowing water using either gamma (Co-60) or proton beam irradiation.

In the heart of our experimental setup is a recirculation loop that is capable of maintaining water inside the irradiation cell (autoclave) at temperatures up to 350°C and pressures up to 200 atm. Essentially, the flow system consists of a feed tank with aqueous solution under study, low and high pressure loops with associated pumps, a preheater, an autoclave and a cooler. Water chemistry control is of a great importance, hence, the automated data acquisition system continuously monitors pressure, temperature, dissolved hydrogen and dissolved oxygen content and water conductivity before and after irradiation. The water chemistry can be easily modified by saturation/doping with the additives of interest, such as hydrogen, oxygen or ionic species.

During water radiolysis a number of radical transients and molecular products are formed, as shown by simplified Equation 1:

$$H_2O \rightarrow e^-_{aq}, \cdot OH,\ \cdot H,\ H^+, \cdot HO_2,\ H_2O_2,\ H_2 \qquad (1)$$

The “yields” of radical species (e^-_{aq},$\cdot$OH, $\cdot$H) are functions of time because their diffusive escape is in competition with recombination processes. At room temperature within about a microsecond the recombination reactions are completed, while the surviving radicals can be measured with appropriate scavengers as an “escape yield”. Scavenged yields of major radical products formed during water radiolysis at room temperature are well established. However, radiation chemical yields and reaction rates of primary radicals in the radiolysis of water above 300°C are either not measured at all or inconsistent; current reactor models include calculations based on extrapolated data [2].

Our first experiments will target radiation chemical yields for the free radicals $\cdot$H and e^-_{aq} [3]. Corresponding G values will be determined for low and high linear energy transfer (LET) types of radiation using, respectively, gamma and ion beam radiolysis. Water radiolysis in a wide range of high temperature and pressure conditions (up to 350°C and 200 atm) will be performed. N_2O and ethanol-d_5 will be used to scavenge e^-_{aq} and $\cdot$H, respectively. Very sensitive mass spectroscopy technique (or its combination with gas chromatography) will be employed to measure N_2, HD, H_2 or O_2 formed either as stable products of water radiolysis (H_2, $H_2O_2 \rightarrow O_2$) or as the products of scavenging reactions ($N_2O \rightarrow N_2$, $C_2D_5OH \rightarrow HD$).

Preliminary data will be used to demonstrate that the described rig can be successfully employed to study free radical processes in aqueous systems at HTHP conditions. The importance of the radiolysis experiments in water at HTHP is hard to underestimate: described studies will produce essential thermodynamic and kinetic data that will aid in development of models relevant to nuclear reactor water chemistry behavior.

References:

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